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**\*\*See application file for complete search history\*\***

REF-CITED:

U.S. PATENT DOCUMENTS

PAT-NO	ISSUE-DATE	PATENTEE-NAME		US-CL
<u>2824620</u>	February 1958	De Rosset	N/A	N/A
N/A				
<u>2848620</u>	August 1958	Backus	N/A	N/A
N/A				
<u>2958391</u>	November 1960	De Rosset	N/A	N/A
N/A				
<u>3022187</u>	February 1962	Eyrand et al.	428/545	N/A
N/A				
<u>3135591</u>	June 1964	Jones	95/53	N/A
N/A				
<u>3208198</u>	September 1965	Rubin	N/A	N/A
N/A				
<u>3241298</u>	March 1966	Pierce	N/A	N/A
N/A				
<u>3245206</u>	April 1966	Bonnet	N/A	N/A
N/A				
<u>3251173</u>	May 1966	Ehlers et al.	N/A	N/A
N/A				
<u>3392510</u>	July 1968	Koch, Jr.	N/A	N/A
N/A				
<u>3413777</u>	December 1968	Langley	N/A	N/A

N/A				
<u>3477288</u>	November 1969	Krcal et al.	N/A	N/A
N/A				
<u>3713270</u>	January 1973	Farr et al.	N/A	N/A
N/A				
<u>3794468</u>	February 1974	Leonard	422/48	N/A
N/A				
<u>4039703</u>	August 1977	Kamijo et al.	N/A	N/A
N/A				
<u>4132660</u>	January 1979	Christian et al.	N/A	N/A
N/A				
<u>4132668</u>	January 1979	Gryaznov et al.	N/A	N/A
N/A				
<u>4472175</u>	September 1984	Malon et al.	N/A	N/A
N/A				
<u>4496373</u>	January 1985	Behr et al.	N/A	N/A
N/A				
<u>4589891</u>	May 1986	Iniotakis et al.	N/A	N/A
N/A				
<u>4689150</u>	August 1987	Abe et al.	N/A	N/A
N/A				
<u>4699637</u>	October 1987	Iniotakis et al.	N/A	N/A
N/A				
<u>4711719</u>	December 1987	Leenaars et al.	N/A	N/A
N/A				
<u>4857080</u>	August 1989	Baker et al.	N/A	N/A
N/A				
<u>4865630</u>	September 1989	Abe	96/11	N/A
N/A				
<u>4880441</u>	November 1989	Kesting et al.	N/A	N/A
N/A				
<u>4971696</u>	November 1990	Abe et al.	N/A	N/A
N/A				
<u>5015269</u>	May 1991	Garrett et al.	N/A	N/A
N/A				
<u>5139540</u>	August 1992	Najjar et al.	N/A	N/A
N/A				
<u>5139541</u>	August 1992	Edlund	N/A	N/A
N/A				
<u>5205841</u>	April 1993	Vaiman	N/A	N/A
N/A				
<u>5217506</u>	June 1993	Edlund et al.	N/A	N/A

N/A				
<u>5259870</u>	November 1993	Edlund	N/A	N/A
N/A				
<u>5310414</u>	May 1994	Najjar et al.	N/A	N/A
N/A				
<u>5342431</u>	August 1994	Anderson et al.	<u>95/45</u>	N/A
N/A				
<u>5358553</u>	October 1994	Najjar et al.	N/A	N/A
N/A				
<u>5376167</u>	December 1994	Broutin et al.	N/A	N/A
N/A				
<u>5384101</u>	January 1995	Rockenfeller	N/A	N/A
N/A				
<u>5393325</u>	February 1995	Edlund	N/A	N/A
N/A				
<u>5415891</u>	May 1995	Liu et al.	N/A	N/A
N/A				
<u>5451386</u>	September 1995	Collins et al.	423/237	N/A
N/A				
<u>5468283</u>	November 1995	French et al.	N/A	N/A
N/A				
<u>5487774</u>	January 1996	Peterson et al.	N/A	N/A
N/A				
<u>5498278</u>	March 1996	Edlund	N/A	N/A
N/A				
<u>5518530</u>	May 1996	Sakai et al.	N/A	N/A
N/A				
<u>5599383</u>	February 1997	Dyer et al.	N/A	N/A
N/A				
<u>5738708</u>	April 1998	Peachey et al.	N/A	N/A
N/A				
<u>5782959</u>	July 1998	Yang et al.	N/A	N/A
N/A				
<u>5782960</u>	July 1998	Ogawa et al.	N/A	N/A
N/A				
<u>5810912</u>	September 1998	Akiyama et al.	N/A	N/A
N/A				
<u>5827569</u>	October 1998	Akiyama et al.	N/A	N/A
N/A				
<u>5891222</u>	April 1999	Hilgendorff et al.	N/A	N/A
N/A				
<u>5904754</u>	May 1999	Juda et al.	N/A	N/A

N/A				
<u>5931987</u>	August 1999	Buxbaum	N/A	N/A
N/A				
<u>5938822</u>	August 1999	Chen et al.	N/A	N/A
N/A				
<u>5955044</u>	September 1999	Lucht et al.	N/A	N/A
N/A				
<u>5980989</u>	November 1999	Takahashi et al.	N/A	N/A
N/A				
<u>5989319</u>	November 1999	Kawae et al.	N/A	N/A
N/A				
<u>5997614</u>	December 1999	Tuma et al.	96/4	N/A
N/A				
<u>6010560</u>	January 2000	Witzko et al.	N/A	N/A
N/A				
<u>6066592</u>	May 2000	Kawae et al.	N/A	N/A
N/A				
<u>6083297</u>	July 2000	Valus et al.	N/A	N/A
N/A				
<u>6083390</u>	July 2000	Hartmann	N/A	N/A
N/A				
<u>6139810</u>	October 2000	Gottzmann et al.	N/A	N/A
N/A				
<u>6152987</u>	November 2000	Ma et al.	N/A	N/A
N/A				
<u>6171574</u>	January 2001	Juda et al.	N/A	N/A
N/A				
<u>6171712</u>	January 2001	Thornton	N/A	N/A
N/A				
<u>6183542</u>	February 2001	Bossard	N/A	N/A
N/A				
<u>6183543</u>	February 2001	Buxbuam	N/A	N/A
N/A				
<u>2001/0013272</u>	August 2001	Blase et al.	96/8	N/A
N/A				

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ABSTRACT:

An improved metal gas separation membrane for separating hydrogen from a gas steam includes a quantity of metal particles that are bonded together to form a porous body. The porous body may have a porosity that increases from a first surface to an opposite second surface and may additionally include a coating of ceramic particles on the first surface. The metal gas separation membrane may include a coating of a dense precious metal applied thereto that is permeable by hydrogen via chemisorption-dissociation-diffusion. The porous body may include a catalytic enhancement. Also disclosed are three gas separation modules that employ the metal gas separation membrane disposed within a core of the gas separation module for separating hydrogen from a gas stream. The gas separation membranes are each supported on a first mounting member and a second mounting member. The gas separation modules may also include a catalytic enhancement.

18 Claims, 8 Drawing figures

Exemplary Claim Number: 1

Number of Drawing Sheets: 4

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Abstract Text - ABTX (1):

An improved metal gas separation membrane for separating hydrogen from a gas steam includes a quantity of metal particles that are bonded together to form a porous body. The porous body may have a porosity that increases from a first surface to an opposite second surface and may additionally include a coating of ceramic particles on the first surface. The metal gas separation membrane may include a coating of a dense precious metal applied thereto that is permeable by hydrogen via chemisorption-dissociation-diffusion. The porous body may include a catalytic enhancement. Also disclosed are three gas separation modules that employ the metal gas separation membrane disposed within a core of the gas separation module for separating hydrogen from a gas stream. The gas separation membranes are each supported on a first mounting member and a second mounting member. The gas separation modules may also include a catalytic enhancement.

TITLE - TI (1):

Metal gas separation membrane module design

Parent Case Text - PCTX (2):

This application is a continuation-in-part application of commonly assigned U.S. patent application Ser. No. 09/822,927 filed Mar. 30, 2001, and entitled "Metal Gas Separation Membrane".

Brief Summary Text - BSTX (3):

The present invention relates generally to gas separation modules and, more particularly, to a gas separation module for the separation of hydrogen gas from a gas stream.

Brief Summary Text - BSTX (5):

Membranes and membrane modules for the separation of hydrogen from other gases are generally known. In particular, useful membranes for hydrogen separation typically can be categorized as being of four general types: (i) polymeric, (ii) porous inorganic, (iii) self-supporting non-porous metal, and (iv) non-porous metal supported on a porous rigid matrix such as metal or ceramic.

Brief Summary Text - BSTX (6):

Porous inorganic-based membranes are typically fabricated from titania, zirconia, alumina, glass, molecular sieving carbon, silica and/or zeolites. All are fabricated with a narrow pore size distribution, with the porous inorganic membranes exhibiting high hydrogen permeability but low selectivity due to relatively large mean pore diameters. Such materials are brittle and thus susceptible to failure due to cracking, and the sealing and fixturing of such porous inorganic-based membranes limit their use to relatively low temperature applications.

Brief Summary Text - BSTX (7):

Development of supported metal membranes has focused on the utilization of ceramic tubes coated with a thin adherently bonded film or foil of non-porous or dense palladium (Pd) or Pd-alloys. The ceramic support tube typically is of a graded porosity from one surface thereof to a second opposite surface. More specifically, the porosity of the ceramic support tube typically is at its least at the surface upon which the Pd or Pd-alloy is disposed, and the porosity of the tube increases from this surface to a maximum porosity on the surface opposite the layer of Pd. The layer of Pd or Pd-alloy is selectively permeable to hydrogen gas and is typically capable of withstanding temperatures of 1500-1600.degree. F. (815-870.degree. C.).

Brief Summary Text - BSTX (8):

Such ceramic-supported metal membranes are typically housed in shell and tube modules and are fitted with compression gaskets to seal the membrane tube

into the module to prevent leakage of the feed gas stream into the permeate gas stream. Potential leak paths between the feed and permeate gas streams can exist due to differences in the coefficients of thermal expansion of the ceramic tube and the metal compression fittings. Additionally, the ceramic support tubes are inherently brittle and can experience long-term thermal fatigue due to repetitive process or system start-up and shutdown cycles.

Brief Summary Text - BSTX (9):

The mechanical adherence of the thin Pd or Pd-alloy layer upon the surface of the ceramic support tube requires secure attachment of the film or foil onto the surface of the ceramic, as well as the absence of pinholes or other mechanical rupturing that can occur during manufacture or use of the ceramic tube membrane.

Brief Summary Text - BSTX (10):

For porous metal membranes such as porous stainless steel, Knudsen diffusion or combined Knudsen diffusion/surface diffusion are the primary mechanisms by which gas transport occurs across the membrane. For dense metal membranes such as Pd or Pd-alloy foil or film, however, the primary mechanism of gas transport through the metal layer is traditional chemisorption-dissociation-diffusion. Broadly stated, chemisorption-dissociation-diffusion transport involves chemisorption of hydrogen molecules onto the membrane surface, dissociation of hydrogen atoms into protons and electrons, transportation of the protons and electrons through the dense metal, reassociation of the protons and electrons into hydrogen molecules, and desorption of hydrogen molecules from the media. While Knudsen diffusion typically offers greater flow rates across a membrane than chemisorption-dissociation-diffusion, Knudsen diffusion suffers from reduced hydrogen selectivity as compared with chemisorption-dissociation-diffusion.

Brief Summary Text - BSTX (11):

It is also known that the interaction of a gas stream with catalytic materials can increase the concentration of hydrogen within the reactant or process gas stream. Such catalytic materials enhance the water gas shift reaction whereby carbon monoxide is reacted with water to form carbon dioxide and hydrogen gas. Catalytic materials also promote the decomposition of ammonia which also increases the concentration of hydrogen.

Brief Summary Text - BSTX (12):

Examples of such catalytic materials include platinum (Pt), palladium (Pd), rhodium (Rh), ruthenium (Ru), nickel (Ni), iridium (Ir), and the like. While it has been known to apply such catalytic materials to ceramic support



substrates to form composite membranes, such composite membranes still suffer from the aforementioned problems associated with the application of Pd and Pd-alloy foils and films to ceramic support tubes.

Brief Summary Text - BSTX (13):

The problems inherent in forming a highly gas specific membrane that is capable of withstanding elevated temperatures are similarly inherent in designing a module that incorporates such membranes. In order to maintain separation between a feed gas stream and a permeate gas stream, it is necessary to provide seals or sealing structures that effectively isolate the feed gas stream from the permeate gas stream. Mechanical and thermal stresses resulting from different coefficients of thermal expansion and elevated temperatures greatly impair the ability of most known sealing systems to effectively isolate the feed gas stream from the permeate gas stream.

Brief Summary Text - BSTX (14):

It is thus desired to provide a hydrogen gas separation membrane and module having high hydrogen selectivity that overcomes the aforementioned problems associated with the application of Pd and Pd-alloys to ceramic support tubes. It is also desired to provide such a membrane and module that include a catalytic enhancement incorporated therein.

Brief Summary Text - BSTX (16):

In view of the foregoing, an improved gas separation module for separating hydrogen from a gas steam includes a metal gas separation membrane having a quantity of metal particles in the form of fibers and/or powders that are compacted together and bonded with one another to form a porous body that is selectively permeable to hydrogen. The porous body may be of a constant porosity throughout or may have a porosity that is "graded," i.e., increases from a first surface to a second opposite surface. A porous body having a graded porosity advantageously provides a low porosity (and thus consequent higher hydrogen selectivity) at the first surface without the higher pressure drop that would be experienced by the hydrogen from the second surface to the first surface if the porous body had the same low porosity throughout its cross section. This variation in porosity can result from the use of metal particles in the form of fibers and/or powders that gradually increase in size in going from the first surface to the second surface, or may result from the use of the same size particles that are compacted to a relatively lesser degree in going from the first surface to the second surface.

Brief Summary Text - BSTX (17):

The metal gas separation membrane may additionally include a coating of

ceramic particles on the first surface thereof to further decrease the porosity at the first surface. Alternatively, or in addition thereto, the metal gas separation membrane may include a thin adherently bonded foil or coating of a dense precious metal such as palladium, palladium-alloys, and the like applied thereto that is permeable by hydrogen according to a chemisorption-dissociation-diffusion transport phenomenon.

Brief Summary Text - BSTX (18):

Still alternatively, or in addition thereto, the gas separation module may include a catalytic enhancement that can interact with a feed gas stream to increase the concentration or quantity of hydrogen in the feed gas stream according to various catalytic reactions such as the water gas shift reaction and the ammonia decomposition reaction. The catalytic enhancement can be in the nature of catalytic materials that are dispersed throughout the porous body, that are supported on scaffold apparatuses such as metal fibers and chemical vapor infiltration silicon carbide (CVI-SiC) reticulated foam, that are coated on the metal particles that make up the porous body, that are in the form of a layer on a surface of the porous body with or without the addition of a ceramic washcoat therebetween, or can be in the form of an application of perovskite, zeolite, or spinel structures to the porous body.

Brief Summary Text - BSTX (19):

The gas separation module includes a plurality of metal gas separation membranes and a housing having first and second mounting members in the form of tube sheets. The metal gas separation membranes are supported on the first and second mounting members. The metal gas separation membranes can advantageously be fixedly mounted, such as by being welded, to the tube sheets to sealingly mount the metal gas separation members to the tube sheets.

Drawing Description Text - DRTX (2):

FIG. 1 is a schematic cross-sectional view of a first embodiment of a gas separation membrane in accordance with the present invention;

Drawing Description Text - DRTX (4):

FIG. 2 is a schematic cross-sectional view of a second embodiment of a gas separation membrane in accordance with the present invention;

Drawing Description Text - DRTX (5):

FIG. 3 is a schematic cross-sectional view of a third embodiment of a gas separation membrane in accordance with the present invention;

Drawing Description Text - DRTX (6):

FIG. 4 is a schematic cross-sectional view of a fourth embodiment of a gas separation membrane in accordance with the present invention;

Drawing Description Text - DRTX (7):

FIG. 5 is a schematic cross-sectional view of a first embodiment of a gas separation module in accordance with the present invention;

Drawing Description Text - DRTX (8):

FIG. 6 is a schematic cross-sectional view of a second embodiment of a gas separation module in accordance with the present invention; and

Drawing Description Text - DRTX (9):

FIG. 7 is a schematic cross-sectional view of a third embodiment of a gas separation module in accordance with the present invention.

Detailed Description Text - DETX (2):

A first embodiment of a metal gas separation membrane 4 in accordance with the present invention is indicated generally in FIG. 1. The metal gas separation membrane 4 is advantageously configured to separate hydrogen gas from a gas stream 8.

Detailed Description Text - DETX (3):

The metal gas separation membrane 4 includes a transmission member 6 and can be of numerous shapes, such as plates, tubes, honeycomb configurations, and other such shapes. The transmission member 6 is depicted schematically in cross-section FIG. 1, and it includes a porous body 10 made out of a plurality of metal particles 12 compressed and bonded together and a metal coating 16 on the porous body 10. The porous body 10 includes a first surface 20 and a second surface 24 opposite one another with the metal coating 16 being disposed on the first surface 20.

Detailed Description Text - DETX (4):

The metal particles 12 that make up the porous body 10 can be of numerous physical configurations such as metal fibers, metal powder, and other shapes, and the porous body 10 can be made up of one type of particle or combinations of these different-shaped metal particles 12. FIG. 1A schematically depicts an example of a portion of a porous body 10 that includes a quantity of metal fibers 25. The metal particles 12 can be fabricated from known metals, superalloys, and/or intermetallic materials to permit the porous body 10 to withstand high temperatures, meaning not only that the material does not melt at the elevated temperatures but also that the material resists corrosion in the potentially oxidizing or reducing environment in which the membrane 4 is

used.

Detailed Description Text - DETX (5):

The metal particles 12 are compacted and sinter bonded to form a thin, dense member of material that makes up the **porous** body 10. The resultant **porous** body 10 has a thickness that is in the range of about 100 microns to 5 millimeters, and preferably in the range of about 0.5 millimeters (500 microns) to 2 millimeters, although other thicknesses may be appropriate depending upon the specific needs of the particular application. In the circumstance where the **porous** body 10 is an elongated substantially cylindrical member formed with an elongated void, the outer diameter of the **porous** body 10 may be in the range of about 5 to 15 millimeters, and preferably is in the range of about 5 to 8 millimeters.

Detailed Description Text - DETX (6):

As can be seen in FIG. 1, the porosity of the **porous** body 10 is greater at the second surface 24 than at the first surface 20, meaning that the space between adjacent metal particles 12 near the second surface 24 is greater than the space between adjacent metal particles 12 near the first surface 20. Alternatively, the **porous** body 10 can be said to have a density that decreases in a direction from the first surface 20 toward the second surface 24. The **porous** body 10 depicted in FIG. 1 thus can be said to have a graded porosity.

Detailed Description Text - DETX (7):

As will be set forth more fully below, the **porous** body 10 is advantageously configured to provide relatively high hydrogen selectivity at the first surface 20 due to its relatively low porosity there. The regions of the **porous** body extending from the first surface 20 to the second surface 24, being relatively more **porous** than the first surface 20, have a relatively lower hydrogen selectivity but correspondingly permit the relatively free flow of **gases** therethrough. These relatively **porous** regions of the **porous** body 10 can thus be said to provide support to the low-porosity region of the **porous** body 10 at the first surface 20, which generally is relatively thin and of lower strength, without meaningfully impeding the flow of hydrogen therethrough from the second surface 24 to the first surface 20. As such, the graded porosity feature of the **porous** body 10 itself provides both high hydrogen selectivity and high hydrogen flow rates therethrough without the high pressure drop that would be experienced by the hydrogen if the porosity of the **porous** body 10 were that of the first surface 20 throughout the **porous** body 10.

Detailed Description Text - DETX (9):

As is shown in FIG. 1, the metal particles 12 are all of substantially the

same size, and the variation in porosity between the first and second surfaces 20 and 24 results from compacting the metal particles 12 to a greater degree at the first surface 20 than at the second surface 24. In this regard, the porous body 10 can be manufactured out of several layers of metal particles 12, with each layer being compacted to a different degree, or other appropriate methodologies can be employed such as forming each layer out of a slurry of metal particles that are sequentially sprayed, painted, or centrifuged onto a surface, with the layers being sprayed, painted, or centrifuged to different degrees.

Detailed Description Text - DETX (10):

The metal coating 16 is a thin dense adherently bonded layer or foil of a precious metal such as palladium (Pd), palladium alloys, and the like that enhance gas phase chemisorption-dissociation-diffusion of hydrogen therethrough. Such palladium alloys may include palladium--copper (Pd--Cu), palladium--ruthenium (Pd--Ru), and other alloys. As is known in the relevant art, the expression gas phase "chemisorption-dissociation-diffusion" of hydrogen refers to molecular chemisorption and dissociation of hydrogen along the high pressure side of the metal coating 16, proton and electron diffusion through the lattice of the metal coating 16, and proton and electron reassociation and recombination and desorption of molecular species along the opposite side of the metal coating 16. The metal coating 16 thus can be referred to as a chemisorption-dissociation-diffusion coating. The metal coating 16 can be applied to the porous body 10 in any of a variety of fashions such as via electroless plating, electroless plating with osmosis, electroplating, sputtering, electrodeposition, and the like.

Detailed Description Text - DETX (11):

In the embodiment depicted in FIG. 1, the metal gas separation membrane 4 receives the gas stream 8 against the second surface 24 and separates hydrogen from the gas stream 8 to form a permeate stream 26 of high purity hydrogen gas that flows out of the side of the metal coating 16 opposite the porous body 10. In alternate embodiments (not depicted,) the overall gas flow may be in a reverse direction to that depicted generally in FIG. 1, i.e., the gas stream 8 may flow directly against the metal coating 16 such that the permeate stream 26 would flow out of the second surface 24.

Detailed Description Text - DETX (12):

By way of example, the metal particles 12 may be in the form of fibers selected to be of a size in the range of about <1 to 50 microns in diameter and generally less than 10 millimeters in length and are compacted to an appropriate degree to permit the flow of gases therethrough. Hydrogen that

passes through the porous body 10 and reaches the first surface 20 is then permitted to flow through the metal coating 16 primarily by chemisorption-dissociation-diffusion.

Detailed Description Text - DETX (14):

It can thus be seen that the porous body 10 serves to mechanically support the metal coating 16 thereon. In this regard, it can be seen that the metal particles 12 provide numerous points of contact between the porous body 10 and the metal coating 16, which helps the metal coating 16 to adhere onto the porous body 10 during operation of the metal gas separation membrane 4 and during the thermal expansion and contraction of the metal gas separation membrane 4 during start-up and shut-down operations.

Detailed Description Text - DETX (15):

In its simplest form, the metal gas separation membrane could comprise solely a porous body being manufactured out of metal particles and having a constant density and porosity throughout, with the porous body serving as the transmission member that separates hydrogen from a gas stream. In addition, the porous body may be of a graded density to reduce the pressure drop across the porous body 10 and/or can additionally include the metal coating 16 to further increase hydrogen selectivity.

Detailed Description Text - DETX (16):

A second embodiment of a metal gas separation membrane 104 is indicated generally in FIG. 2. The metal gas separation membrane 104 includes a transmission member 106 having a porous body 110 in which the graded porosity thereof is achieved by providing metal particles 112 that vary in size from relatively finer metal particles 112A at the first surface 120 to relatively coarser metal particles 112B at the second surface 124.

Detailed Description Text - DETX (18):

A third embodiment of a metal gas separation membrane 1004 in accordance with the present invention is indicated generally in FIG. 3. The metal gas separation membrane 1004 is similar to the metal gas separation membranes set forth above and includes a porous body 1010 having a first surface 1020 and a second surface 1024 opposite one another, a layer of ceramic particles 1028 disposed on the first surface 1020, and a chemisorption-dissociation-diffusion coating 1016 disposed on the layer of ceramic particles 1028. The porous body 1010 is of a graded porosity that results from the use of metal particles of varying sizes that range from relatively finer metal particles 1012A disposed at the first surface 1020 to relatively coarser metal particles 1012B disposed at the second surface 1024.

Detailed Description Text - DETX (19):

The metal gas separation membrane 1004 may be of a hollow, generally cylindrical configuration, although other configurations such as those set forth above, may be employed without departing from the concept of the present invention. In such a hollow cylindrical configuration, the outer diameter of the metal gas separation membrane 1004 may be in the range of about 5 to 15 millimeters, and preferably in the range of about 5 to 8 millimeters, although the outer diameter may be larger or smaller than that set forth herein. The wall thickness of the porous body 1010 may be in the range of about 0.1 millimeters (100 microns) to 5 millimeters, and preferably in the range of about 0.5 millimeters (500 microns) to 2 millimeters, and such wall thickness is generally defined as the distance between the first surface 1020 and the second surface 1024.

Detailed Description Text - DETX (22):

The porous body 1010 may be made up of known materials such as FeAl, Haynes.RTM. 160 (available from Haynes International of Kokomo, Ind. 46904, USA), RA-333.RTM. or RA85H.RTM. (available from Rolled Alloys, Temperance, Mich. 48182, USA), 602CA, and Fecralloy, although other materials including those set forth above may be employed. The size range of the finer and coarser metal particles 1012A and 1012B may be the same as that set forth above or may be of other sizes depending upon the specific needs of the particular application.

Detailed Description Text - DETX (23):

A fourth embodiment of a metal gas separation membrane 1104 in accordance with the present invention is indicated generally in FIG. 4. The metal gas separation membrane 1104 is similar to the metal gas separation 1004 but includes a porous body 1110 having a catalytic enhancement in the form of particles of catalytic material that range in size from relatively finer particles of catalytic material 1118A disposed at the first surface 1120 to relatively coarser particles of catalytic material 1118B disposed at the second surface 1124 in a fashion similar to the relatively finer and coarser metal particles 1112A and 1112B. The particles of catalytic material 1118A and 1118B are particles of precious metals such as platinum (Pt), palladium (Pd), rhodium (Rh), ruthenium (Ru), nickel (Ni), iridium (Ir), and alloys thereof and are depicted in FIG. 4 as being shaded. The particles of catalytic material 1118A and 1118B promote the water gas shift reaction, the ammonia decomposition reaction, and the like in a known fashion to increase the concentration or availability of hydrogen. Alternately, fine catalytic particles of uniform size can be dispersed throughout the porous body 1110 and/or a layer of ceramic

particles 1128 disposed against the first surface 1120, and may coat or be adhered to the metal and/or the ceramic particles. The metal **gas separation membrane** 1104 additionally includes a support member 1144 disposed against the chemisorption-dissociation-diffusion coating 1116. It is understood that the metal **gas separation membranes** 4, 104, and 1004 can be catalytically enhanced in any of the fashions set forth above and/or can include the support member 1144.

Detailed Description Text - DETX (24):

The metal **gas separation membrane** 1104 may be employed in an environment in which a **gas** stream flows against or along the second surface 1124 and into the **porous** body 1110 where it catalytically interacts with the particles of catalytic material 1118A and 1118B. Hydrogen **gas** then flows through the layer of ceramic particles 1128 and passes through the chemisorption-dissociation-diffusion coating 1116 in a fashion set forth above.

Detailed Description Text - DETX (25):

The support member 1144 retains the chemisorption-dissociation-diffusion coating 1116 and the layer of ceramic particles 1128 disposed on the **porous** body 1110 when **gas** flows through the **porous** body 1110 prior to contacting the chemisorption-dissociation-diffusion coating 1116. In this regard, the support member 1144 helps to resist the chemisorption-dissociation-diffusion coating 1116 and the layer of ceramic particles 1128 from being blown away from or becoming otherwise dislodged from the **porous** body 1110 as a result of the substantial pressure drop in going from the second surface 1124 to the outermost portions of the chemisorption-dissociation-diffusion coating 1116. The support member 1144 may be in the form of a metal mesh, a screen, a densified perforated sleeve, as well as other appropriate configurations.

Detailed Description Text - DETX (26):

A first embodiment of a **gas separation** module 1258 in accordance with the present invention is indicated generally in FIG. 5. The **gas separation** module 1258 includes a housing 1260 within which are disposed a plurality of metal **gas separation membranes** 1204 in accordance with the present invention. While the metal **gas separation membranes** 1204 employed in the **gas separation** module 1258 may be the metal **gas separation membranes** 1004, it is understood that any of the metal **gas separation membranes** set forth herein may be employed in the **gas separation** module 1258 without departing from the concept of the present invention.

Detailed Description Text - DETX (27):

In general terms, the **gas separation** module 1258 can be stated as being



structured to receive a feed gas stream 1208 and to separate it into a permeate gas stream 1226 and a retentate gas stream 1261. While the permeate gas stream 1226 may be high purity hydrogen gas, it is understood that the gas separation module 1258 potentially could be configured such that the permeate gas stream 1226 is another gas such as oxygen.

Detailed Description Text - DETX (29):

Each metal gas separation membrane 1204 is an elongated substantially cylindrical member having a first end 1268 and an opposite second end 1270 and being formed with an elongated void 1274 formed therein and extending substantially throughout the longitudinal extent of the metal gas separation membrane 1204. Each metal gas separation membrane 1204 includes a first surface 1220 and a second surface 1224, with the second surface 1224 being defined by the void 1274. For purposes of clarity, the metal gas separation membranes 1204 shown in FIG. 5 do not depict a layer of ceramic particles, a chemisorption-dissociation-diffusion coating, or a support member, although it is understood that one or more of these features may be incorporated therein.

Detailed Description Text - DETX (30):

In accordance with the present invention, the metal gas separation membranes 1204 are supported on the first and second mounting members 1264 and 1266. More specifically, the metal gas separation membranes 1204 are fixedly attached to the first and second mounting members 1264 and 1266 such as by welding or other appropriate attachment methodology.

Detailed Description Text - DETX (31):

By configuring the permeable bodies of the metal gas separation membranes 1204 to be formed from a trace of ceramic or other metallic oxide, the metal gas separation membranes 1204 can be welded to the metallic first and second mounting members 1264 and 1266. Such welded attachment is particularly advantageous because it overcomes the problems inherent in attempting to seal a joint between a ceramic member and a metallic member such as occurred with previously known ceramic support structures in previously known gas separation membranes. Additionally, the metal gas separation membranes 1204 are less susceptible to thermal expansion than ceramic members, and furthermore are not subject to configuration deformation problems that can plague ceramic members during firing operations.

Detailed Description Text - DETX (32):

As can be seen in FIG. 5, the first ends 1268 of the metal gas separation membranes 1204 are sealed and sealingly mounted to the first mounting member 1264, whereby the first ends 1268 are closed. In contrast thereto, the

second ends 1270 are mounted adjacent holes 1276 formed in the second mounting member 1266, whereby the second ends 1270 are open. The first mounting member 1264 is formed with a plurality of perforations 1278 in the regions thereof that are not used for sealing the first ends 1268 of the metal gas separation membranes 1204.

Detailed Description Text - DETX (33):

The metal gas separation membranes 1204 each include a generally defined interior 1277 and a generally defined exterior 1279. The interior 1277 of each metal gas separation membrane 1204 is defined by the void 1274. The exterior 1279 is opposite the interior 1277 and is defined as being the regions that are disposed radially outwardly beyond the outermost components of the metal gas separation membranes 1204, whether the outermost components be the porous body, a layer of ceramic particles, a chemisorption-dissociation-diffusion coating, or a support member.

Detailed Description Text - DETX (34):

The first and second mounting members 1264 and 1266 define an inlet manifold 1282 therebetween that is disposed to the exterior 1279 of all of the metal gas separation membranes 1204. Stated otherwise, the inlet manifold 1282 is the portion of the core 1262 between the first and second mounting members 1264 and 1266 that is not occupied by the metal gas separation membranes 1204. Similarly, the second mounting member 1266 defines a permeate gas manifold 1284 extending from the side of the second mounting member 1266 opposite the inlet manifold 1282. Still similarly, the first mounting member 1264 defines a retentate manifold 1286 extending from the side of the first mounting member 1264 opposite the inlet manifold 1282. The permeate gas manifold 1284 and the retentate manifold 1286 are disposed within the core 1262.

Detailed Description Text - DETX (35):

The housing 1200 is additionally formed with a feed gas inlet 1288, a retentate gas outlet 1290, and a permeate gas outlet 1292. The permeate gas outlet 1292 is in fluid communication with the permeate gas manifold 1284. The retentate gas outlet 1290 is in fluid communication with the retentate manifold 1286. The feed gas inlet 1288 is in fluid communication with the inlet manifold 1282. Inasmuch as the first mounting member 1264 is formed with the perforations 1278, the feed gas inlet 1288 is additionally in fluid communication with the retentate manifold 1286 and the retentate gas outlet 1290 through the perforations 1278.

Detailed Description Text - DETX (36):

The gas separ 1208 additionally includes a catalytic enhancement

1294. In the embodiment depicted in FIG. 5, the catalytic enhancement 1294 is disposed within the inlet manifold 1282 of the housing 1260, which is generally to the exterior 1279 of the metal gas separation membranes 1204. The catalytic enhancement 1294 is in the form of catalytic materials 1296 that are disposed on or are otherwise supported by a scaffold apparatus 1298. The catalytic materials 1296 may include any of the catalytic materials set forth herein.

#### Detailed Description Text - DETX (38):

In operation, the feed gas stream 1208 flows through the feed gas inlet 1288 and into the inlet manifold 1282 where it flows into contact with the catalytic enhancement 1294. The catalytic enhancement 1294 promotes the water gas shift reaction, the ammonia decomposition reaction, and the like to increase the concentration or the availability of hydrogen within the feed gas stream 1208. The catalytic enhancement 1294 is advantageously configured to be highly porous and thus permit the free flow of the feed gas stream 1208 therethrough. Depending upon the specific makeup of the feed gas stream 1208 or the specific needs of the particular application, the gas separation module 1258 may be configured to not include the catalytic enhancement 1294.

#### Detailed Description Text - DETX (39):

The feed gas stream 1208 including gaseous hydrogen thereafter flows into contact with the metal gas separation membranes 1204. Hydrogen gas flows through the metal gas separation membranes 1204 from the exterior 1279 thereof to the interior 1277 thereof along the voids 1274 to form the permeate gas stream 1226 which then flows into the permeate gas manifold 1284 and out of the permeate gas outlet 1292. The feed gas stream 1208 from which the hydrogen has been substantially removed flows through the perforations 1278 and into the retentate manifold 1286 where it forms the retentate gas stream 1261 that flows out of the retentate manifold 1286.

#### Detailed Description Text - DETX (40):

The perforations 1278 formed in the first mounting member 1264 help to retain the catalytic enhancement 1294 within the inlet manifold 1282 and additionally assist in managing the substantial pressure drop between the feed gas stream 1208 and the retentate gas stream 1261 that helps to drive the separation of the permeate gas stream 1226 from the feed gas stream 1208. Since the metal gas separation membranes 1204 and the first and second mounting members 1264 and 1266 are manufactured substantially out of metal, the metal gas separation membranes 1204 can be welded to the first and second mounting members 1264 and 1266 which sealing attachment additionally makes the gas separation module 1258 well suited to the elevated temperatures and pressures typically encountered in separating the permeate gas stream 1226 from

## EAST Search History

Ref #	Hits	Search Query	DBs	Default Operator	Plurals	Time Stamp
L1	11570	inorganic membrane and gas separation	USPAT; EPO; DERWENT	AND	ON	2006/05/12 20:53
L2	65793	1 and carbon or coke	USPAT; EPO; DERWENT	AND	ON	2006/05/12 20:53
L3	6871	2 and zeolite	USPAT; EPO; DERWENT	AND	ON	2006/05/12 20:54
L4	3074	3 and porous	USPAT; EPO; DERWENT	AND	ON	2006/05/12 20:55
L5	1239	4 and zeolite same carbon	USPAT; EPO; DERWENT	AND	ON	2006/05/12 21:03
L6	54	95/45 and 4	USPAT; EPO; DERWENT	AND	ON	2006/05/12 21:01
L7	4	6 and coke	USPAT; EPO; DERWENT	AND	ON	2006/05/12 21:01
L8	997	5 and coke	USPAT; EPO; DERWENT	AND	ON	2006/05/12 21:03
L9	3200	coke same hydrocarbon and zeolite	USPAT; EPO; DERWENT	AND	ON	2006/05/12 21:04
L10	10	9 and "coating coke"	USPAT; EPO; DERWENT	AND	ON	2006/05/12 21:04

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